Studying Nanomagnetism with Computational Techniques

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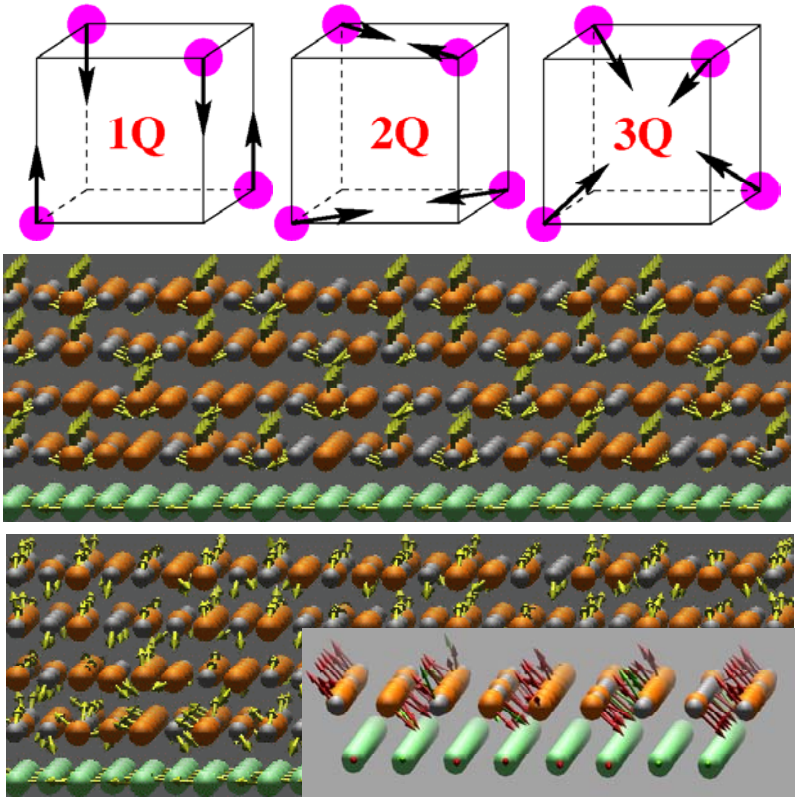
It is well established from decades of research in magnetism and magnetic materials that interface magnetism can be quite complex and different from bulk magnetism. Due to reduced symmetry, the magnetic anisotropy can be orders of magnitude larger than in the bulk. This can lead to considerable frustration and, in some cases, reorientation of the magnetization at the surface and interface. Interfaces between dissimilar materials can change their properties. For example, when in contact with an antiferromagnet, the properties of a ferromagnet change dramatically, the coercive field is usually much larger and, in many cases, the magnetization curve becomes asymmetric showing the exchange bias effect. At the nanoscale, interface effects are expected to be even more significant. Straightforward geometric reasoning shows that the interface region is an important part of the entire structure. For example, in a FePt nanoparticle of 6nm diameter, about 40% of the atoms are located within one lattice constant of the surface. Surface segregation and surface magnetism are expected to have a dominant effect on the magnetic state of such a particle. A few cases that show how dominant surface effects can be in nanostructures already exist in the literature. For example, Kodama *et al.*[i] found that the dramatic increase in coercive field of $FeNi_2O_4$ nanoparticles over the bulk system is due to a spin-glass like surface spin structure to which the ferimagnetic core of the particle couples. Understanding the complex spin structure of magnetic nanostructures at the atomic scale is thus essential for the understanding of nanomagnetism itself. Measuring the non-collinear magnetic state of a nanostructure is a difficult task, since most experimental probes for magnetism average over length scales of ~100 nm or more. Information about the atomic scale spin structure is usually inferred by comparison with model calculations. In many nano-scale systems, however, calculations cannot be based on bulk models, since parameters such as exchange and anisotropy constants, as well as composition profile and atomic valence, are no longer similar to their bulk value. Model calculations, therefore, have to be based on first principles techniques that require only input of the atomic number and are capable of predicting non-collinear magnetic states, dynamic of atomic moments, and response to external stimuli.

Methods to calculate the electronic structure of magnetic nanosystems are mostly based on the local spin-density approximation (LSDA) to Density Functional Theory[ii] (DFT). These have proven very reliable in predicting ground state properties such as magnetic moments and anisotropies[iii], as well as the temperature of magnetic phase transitions[iv]. Extensions of the LSDA to study the dynamics[v] of non-collinear spin systems (in particular constrained LSDA[vi]) are now available. They have been successfully applied to predict non-collinear spin structures of complex antiferromagnets such as FeMn [cite] as well as the role of induced moments on highly susceptible elements such as Pt, Pd, or Ru on the magnetic exchange and anisotropy in important systems such as FePt, CoPt, and FeRu[vii], as well as reorientation transitions in thin films[viii].



Upper panel: three spin states consistent with neutron scattering measurements of γ FeMn. Mössbauer spectroscopy indicates that the 1Q state is not present first principles calculations show that the 2Q and 3Q states are the only stable states where the latter is energetically the most favorable.
Middle panel: starting spins configuration for a first principles calculation of a FeMn/Co multilayer. Co (green) is assumed to be in a ferromagnetic state while FeMn is initiated in a perfect 3Q state.
Lower panel: final configuration after full relaxation of the magnetic state. Co atoms are still perfectly ferromagnetically ordered. The spin structure of FeMn has rearranged and resembles a 1Q state with Fe and Mn moments aligned perpendicular to the Co moments.

In recent years, the development of order *N* electronic structure methods [LSMS] and their implementation on modern high performance computing hardware [cite], have made possible simulations of non-collinear spin systems with thousands of atoms in the unit cell. With these capabilities, it was possible to show [cite], that the spin-structure of FeMn in a Co/FeMn thin film heterostructure reorients from the bulk 3Q structures into a quasi 1Q structure in the thin film with preferential moment directions perpendicular to the ferromagnetic Co moments (see Figure). Present day calculations are already large enough to allow prediction of magnetic ground state properties of nanostructures. It is expected that computer performance will increase by a factor of 100 to 1000 in the next five to ten years. With these capabilities, it will be possible to study finite temperature properties and dynamics of the magnetic state in nanostructures. As a specific example, we consider nanoparticles: particles with 5-10 nm diameter consist of 5,000-20,000 atoms. On vector computers with ~10^4 fast vector processors, it will be possible to integrate the Landau Lishitz equation in which the effective fields and the magnetic moments are computed in first principles electronic structure calculations. On massively parallel computers with >10^5 super scalar processors, it will be possible to calculate temperature dependent magnetic free energies of nano-particles with ab initio Monte Carlo techniques. In these approaches, the energies of individual spin configurations is calculated with first principles electronic structure methods and novel Monte Carlo techniques are used to sample configuration space, thus allowing the calculations of the entropic part of the free energy. Incorporating temperature and dynamics effects in the predictions of magnetic properties is crucial for nanostructures, since the effects of fluctuations at the atomic scale are important for nanomagnetism.

References:

[i] Kodama PRL and JMMM review.
[ii] DFT books
[iii] Kübler book on itinerant magnetism
[iv] Staunton PRL
[v] Antropov PRL
[vi] Mryasov PRB 92 and Stock Phil Mag
[vii] Mryasov JMMM and TBP, Brown, PRB 1993
[viii] Vienna group papers